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Geometric Frustration in the Mixed Layer Pnictide Oxides

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Geometric frustration in the mixed layer pnictide oxides

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We present results from a Monte Carlo investigation of a simple bilayer model with geometrically frustrated interactions similar to those found in the mixed layer pnictide oxides ($Sr_2Mn_3Pn_2O_2, Pn = As, Sb$). Our model is composed of two inequivalent square lattices with nearest-neighbor intra- and interlayer interactions. We find a ground state composed of two independent Néel ordered layers when the interlayer exchange is an order of magnitude weaker than the intralayer exchange, as suggested by experiment. Evidence for local orthogonal order between the layers is found, but it occurs in regions of parameter space which are not experimentally realized. Qualitatively similar results were observed in models with a larger number of layers. We conclude that frustration caused by nearest–neighbor interactions in the mixed layer pnictide oxides is not sufficient to explain the long–range orthogonal order that is observed experimentally.

I. INTRODUCTION

Clean systems of interacting moments have been studied extensively by analytic and numerical techniques. Although, simplified models like Ising, Heisenberg, and Hubbard only retain the most fundamental interactions observed in real materials, they remain tractable to current theoretical techniques, and the study of their ordered phases in various regions of parameter space has contributed enormously to our understanding of magnetic phenomena and the physics of correlated systems. [1] However, real materials are never clean. There is often frustration due to competing interactions and disorder in the interaction strengths.

Competing interactions that cause magnetic frustration can have many origins, lattice geometry, magnetic and non-magnetic impurities. In three dimensions, helical magnetic order has been observed when geometric frustration is accompanied by anisotropy. [2,3] It has also been suggested that some non-collinear spin ordered structures belong to a new chiral universality class. [4] Spin glasses phases are observed when frustration is accompanied by random disorder. [5–7] The systems we study are essentially two dimensional, contain no anisotropic terms or disorder, and frustration is caused by the lattice geometry. Our primary focus is the orthogonal magnetic structure observed in the mixed layer pnictide oxides. [8] A more complete study of these systems has been reported elsewhere. [9] In this note, we present results from larger systems sizes.

II. MODEL

In the pnictide oxides of type $Sr_2Mn_3Pn_2O_2 (Pn = As, Sb)$, two distinct square plane of manganese exist in a lattice of space group symmetry $I4/mmm$. In one layer, manganese is bonded to oxygen in a planar CuO_2 arrangement, MnO_2^{2-}. In a second layer, it is bonded to a pnictogen in a tetrahedral structure, $MnPn_2^{2-}$, where pnictogen atoms project alternately above and below the plane defined by the manganese atoms. From here on we denote the two manganese layers as $Mn1$ for MnO_2^{2-} and $Mn2$ for $MnPn_2^{2-}$. The manganese atoms from the two planes are arranged such that a site in the $Mn1$ layer sits directly above and below the center of a square plaquette of manganese atoms in the $Mn2$ layer. The manganese carry a spin $S = 5/2$.

The pnictide oxides are layered antiferromagnets, and in the case of $Sr_2Mn_3Sb_2O_2$ there is long range order in the planes that eventually gives rise to weak $3D$ order. The ability of ordered planes to drive c-axis order has been investigate before in the case of layered antiferromagnets. [10–12] In the $Mn1$ layers, magnetic order is established along the a-axis of the magnetic unit cell, while in the $Mn2$ layers the magnetization is along the c-axis. Hence there is an orthogonal alignment between neighboring layers. Such an ordered state is not without precedent. [13,14] With experiments indicating different ordering temperatures for the layers ($T_{Mn2} = 300-340K$ and $T_{Mn1} = 50 - 100K$) and the symmetry of the frustrated interlayer interactions, one might expect a two Néel ordered layers with an arbitrary alignment between the magnetizations. It has been shown, however, that thermal or quantum fluctuations (in frustrated systems) can lift the degeneracy of the system to select a single state. [15,16]

To study the effect of frustration on the ground state magnetic order of the pnictide oxides, we develop a simple model of classical Heisenberg spins with nearest neighbor intra- and interlayer interactions. Our model lattice is formed from two layers, one each of type $Mn1$ and $Mn2$, see Fig. 1 The $Mn2$ layer has a lattice constant $a = 1$ and contains n^2 sites. The $Mn2$ layer is larger by a factor $\sqrt{2}$ and is rotated by $\pi/4$ with respect to the lattice directions of the other layer. The $Mn1$ layer contains $n^2/2 + n + 1$ spins. Frustration enters through the interlayer couplings.

The Hamiltonian for our bilayer model is written as

$$H = J_1 \sum_{i,\vec{\delta}_1} \vec{S}_i^{(1)} \cdot \vec{S}_{i+\vec{\delta}_1}^{(1)} + J_2 \sum_{i,\vec{\delta}_2} \vec{S}_i^{(2)} \cdot \vec{S}_{i+\vec{\delta}_2}^{(2)} + J_\perp \sum_{i,\vec{\delta}_\perp} \vec{S}_i^{(\alpha)} \cdot \vec{S}_{i+\vec{\delta}_\perp}^{(\beta)}. \quad (1)$$

The constants J_1, J_2, and $J_\perp$ represent the $Mn1$ and $Mn2$ intralayer couplings and the interlayer coupling, respectively. The summations of $\vec{\delta}_\mu$ are over nearest neighbors to site i. For classical spins, one has $|\vec{S}| = (S_x^2 + S_y^2 + S_z^2)^{1/2} = 1$. The relatively large spin-5/2 of the Mn atoms in the pnictide oxides makes this a reasonable approximation.

Our principal method to study Eq. 1 is a single spin flip Monte Carlo algorithm. We have addressed concerns about proper sampling of phase space by performing simulations with random and ordered initial configurations. In all cases considered, we observed convergence to a unique solution. We have also considered the effects of the boundary on our finite simulations by employing few different boundary conditions: open, periodic, and periodic with an effective field on the $Mn1$ edge sites. Again, we find no qualitative difference in our results due to the conditions imposed at the boundary.

To determine the relative orientation between neighboring spins, either within the same layer or in different layers, we define a collinear

$$C_\parallel^{\alpha,\beta} = \left\langle \frac{1}{zN_\alpha} \sum_i \sum_{\vec{\delta}} (\vec{S}_i^{\alpha} \cdot \vec{S}_{i+\vec{\delta}}^{\beta})^2 \right\rangle, \quad (2)$$

and a perpendicular

$$C_\perp^{\alpha,\beta} = \left\langle \frac{1}{zN_\alpha} \sum_i \sum_{\vec{\delta}} (\vec{S}_i^{\alpha} \times \vec{S}_{i+\vec{\delta}}^{\beta})^2 \right\rangle. \quad (3)$$

spin–spin correlation function. Summations are performed over all all nearest neighbors $\vec{\delta}$ of site i and then over all sites in the lattice; z is the coordination number and N_α is the number of sites in layer α. Intralayer correlations are denoted by $\alpha = \beta$ and interlayer correlations are represented by $\alpha \neq \beta$. We stress that $C_\parallel$ and $C_\perp$ measure local correlations. For classical Heisenberg spins, these correlations take on the simple forms $C_\parallel = \langle \cos^2\theta \rangle$ and $C_\perp = \langle \sin^2\theta \rangle$. In the high temperature, paramagnetic, limit, the values $C_\parallel = 1/3$ and $C_\perp = 2/3$ are obtained.

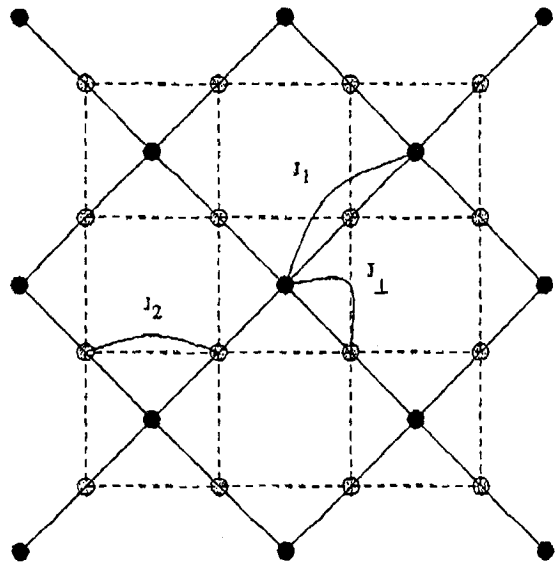

FIG. 1. A 2D projection of the two distinct layers of the pnictice oxide $Sr_2Mn_2Sb_2O_2$. Sites in the $Mn1$ layer are represented by dark circles while sites in the $Mn2$ layer are represented by light circles. The lattice constant for the $Mn2$ plane is set at $a = 1$; therefore, the $Mn1$ square plane is described by the constant $b = \sqrt{2}$. The intralayer couplings are shown as J_1 and J_2, and the interlayer interaction is indicated by $J_\perp$.

III. RESULTS

From the experimental data, representative couplings would set the $Mn2$ intralayer exchange to be stronger than the $Mn1$ intralayer exchange, with the interlayer interaction weaker by at least an order of magnitude. Therefore, experimentally motivated couplings in our model are set to $J_2 = 2.0, J_1 = 1.0$ and $J_\perp = 0.1$. Simulations of such a bilayer model indicate that, as a function of temperature, the moments with in each layer begin to order when the temperature equals the respective energy scale, e.g., $T_{Mn1} = J_1$ and $T_{Mn2} = J_2$. However, the eventual ground state is one of two Néel ordered layers with an arbitrary orientation between the magnetization directions. [9] In a simulation of a four layer system (i.e., a sequence $Mn1 - Mn2 - Mn1 - Mn2$) with periodic boundary conditions along the c-axis, qualitatively the same result is observed, see Fig. 2. We underscore, that the interlayer spin–spin correlation, $C_\parallel^{1,2}$, remains at the paramagnetic limit down to very low temperatures, $T \lesssim J_\perp$.

In simulations where the temperature is fixed and a sweep in $J_\perp$ is performed, two independent layers with an arbitrary alignment was observed for $J_\perp < 0.25$, but

the system moved toward a collinear arrangement between the layers at $J_\perp > 0.4$. We observed this behavior independent of the initial configuration. The transition from a paramagnetic to a collinear interplane state is more abrupt at lower temperatures, see Fig. 3.

An orthogonal state for our bilayer model can be found but at couplings that are not supported by experiments, refer to ref. 9. In the case where $J_2 = 0$, the resultant model is a network of intersecting zigzag chains. Setting $J_\perp = 1$, which acts as the nearest neighbor interaction, and sweeping in J_1, a second neighbor coupling, yields a transition to a uniformly canted state with the behavior of $C_\parallel^{1,2}$ qualitatively similar to that observed for lattice of antiferromagnetically linked zigzag chains. In another case where $J_1 = 0$ and $J_2 = J_\perp$, a Néel ordered $Mn2$ layer results with $Mn1$ spins orthogonal to the local $Mn2$ environment. Turning on J_1 orders the $Mn1$ layer and drives the system to a collinear alignment.

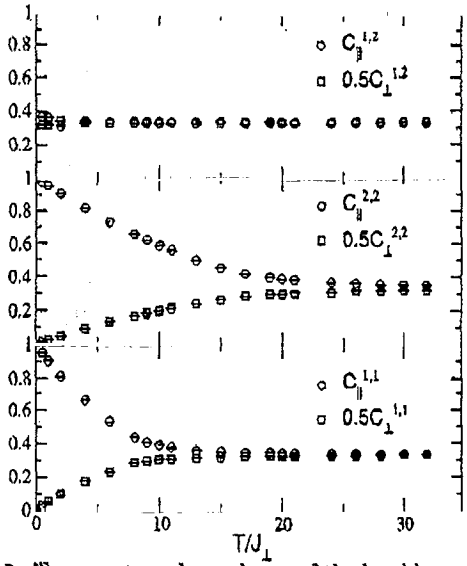

FIG. 2. Temperature dependence of the local intra- and interlayer spin–spin correlations as on $40 \times 40 \times 4$ lattice with periodic boundary conditions and $J_1 = 1.0, J_2 = 2.0, J_\perp = 0.1$. A parallel alignment is favored for intralayer spins when the temperature drops below the respective intralayer coupling; however, the interlayer correlations remain at the high temperature limit of 1/3 even for $T \lessapprox J_\perp$.

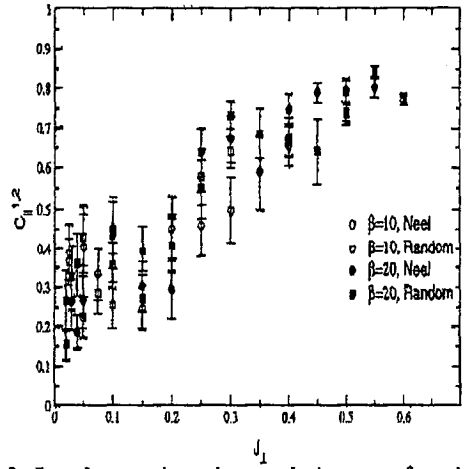

FIG. 3. Interlayer spin–spin correlations as a function of $J_\perp$ with $J_1 = 1.0$ and $J_2 = 2.0$. The simulation was initialized in either a random or Néel state.

IV. CONCLUSIONS

We conclude that frustration caused by nearest neighbor interactions, both intra– and interlayer, in the mixed layer pnictide oxides is not sufficient to explain the long range orthogonal order that is observed experimentally. Thus, in these systems it is likely that other terms in the Hamiltonian, e.g., local anisotropies, are required to explain the magnetic behavior.

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